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Partitioning of oxygen during core formation on the Earth and Mars

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Core formation on the Earth and Mars involved the physical separation of metal and silicate, most probably in deep magma oceans^{1–4}. Although core-formation models explain many aspects of mantle geochemistry, they have not accounted for the large differences observed between the compositions of the mantles of the Earth (~8 wt% FeO) and Mars (~18 wt% FeO) or the smaller

mass fraction of the martian core^{5–7}. Here we explain these differences as a consequence of the solubility of oxygen in liquid iron-alloy increasing with increasing temperature. We assume that the Earth and Mars both accreted from oxidized chondritic material. In a terrestrial magma ocean, 1,200–2,000 km deep, high temperatures resulted in the extraction of FeO from the silicate magma ocean owing to high solubility of oxygen in the metal. Lower temperatures of a martian magma ocean resulted in little or no extraction of FeO from the mantle, which thus remains FeO-rich. The FeO extracted from the Earth's magma ocean may have contributed to chemical heterogeneities in the lowermost mantle⁸, a FeO-rich D'' layer⁹ and the light element budget of the core^{10,11}.

The most significant differentiation event in the history of the Earth and other terrestrial planets was the separation of metal and silicate to form a metallic Fe-rich core and a silicate mantle. The geochemical consequences of this process have been studied extensively in recent years to explain the geochemistry of the Earth's mantle and the physical mechanisms of core formation and accretion. Extensive melting of the Earth and the formation of a deep magma ocean as a consequence of one or more giant impacts probably facilitated metal–silicate separation^{12,13}. The content of moderately siderophile (metal-loving) elements, such as Ni and Co, in the Earth's mantle can be explained by the separation of liquid metal and silicate in a magma ocean at least 700 km deep^{1–3}. This conclusion is based on experimental studies of the partitioning of siderophile elements between liquid metal and liquid silicate at high pressure. Here we consider the partitioning of oxygen between metal and silicate in a magma ocean with the aim of understanding how the FeO content and oxidation state of planetary mantles are affected by core formation.

We studied the solubility of oxygen in liquid Fe–Ni alloy at 9 and 18 GPa, 2,173–2,673 K and oxygen fugacities (f_{O_2}) 1.1 to 3.6 log units below the iron–wüstite buffer by equilibrating the samples with magnesiowüstite in a multi-anvil apparatus. Details of the starting materials and experimental and analytical methods have been described previously¹⁴. Over the range of experimental conditions, the solubility varies from below detection limit to 1.28 wt% (Fig. 1a, Table 1). The results show that oxygen solubility increases with increasing temperature and oxygen fugacity (Fig. 1a). To determine the effects of pressure and temperature independently of f_{O_2} , we calculate the distribution coefficient, K_d , for the partitioning of oxygen between liquid Fe-alloy and magnesiowüstite:

$$K_d = \frac{X_{\text{O}}^{\text{met}} X_{\text{Fe}}^{\text{met}}}{X_{\text{FeO}}^{\text{mw}}} \quad (1)$$

where $X_{\text{O}}^{\text{met}}$, $X_{\text{Fe}}^{\text{met}}$ and $X_{\text{FeO}}^{\text{mw}}$ are the mole fractions of oxygen in metal, Fe in metal and FeO in magnesiowüstite respectively. As shown in Fig. 1b, the distribution coefficient, and therefore the oxygen solubility at constant f_{O_2} , decreases with increasing pressure. These trends are consistent with previous results obtained at 5–25 GPa and 2,073–2,773 K but over a relatively restricted f_{O_2} range¹⁵. Note that the results are contrary to early predictions that the solubility of oxygen in liquid Fe should increase with increasing pressure¹⁶.

To extrapolate the oxygen solubility data as a function of pressure (P) and temperature (T), we use:

$$RT \ln K_d = -\Delta H + T\Delta S - P\Delta V \quad (2)$$

where ΔH , ΔS and ΔV are the changes in enthalpy, entropy and volume, respectively, for the oxygen exchange reaction and R is the gas constant. We fitted equation (2) to the data, including five experimental results obtained up to 25 GPa by ref. 15. Results of the fit, shown in Fig. 1a and b, give $\Delta H = 153,000 (\pm 29,000) \text{ J mol}^{-1}$, $\Delta S = 50.9 (\pm 12.4) \text{ J K}^{-1} \text{ mol}^{-1}$ and $\Delta V = 1,448 (\pm 440) \text{ J GPa}^{-1}$ ($1.448 \pm 0.44 \text{ cm}^3 \text{ mol}^{-1}$).

Extrapolations of the solubility results to higher pressures and

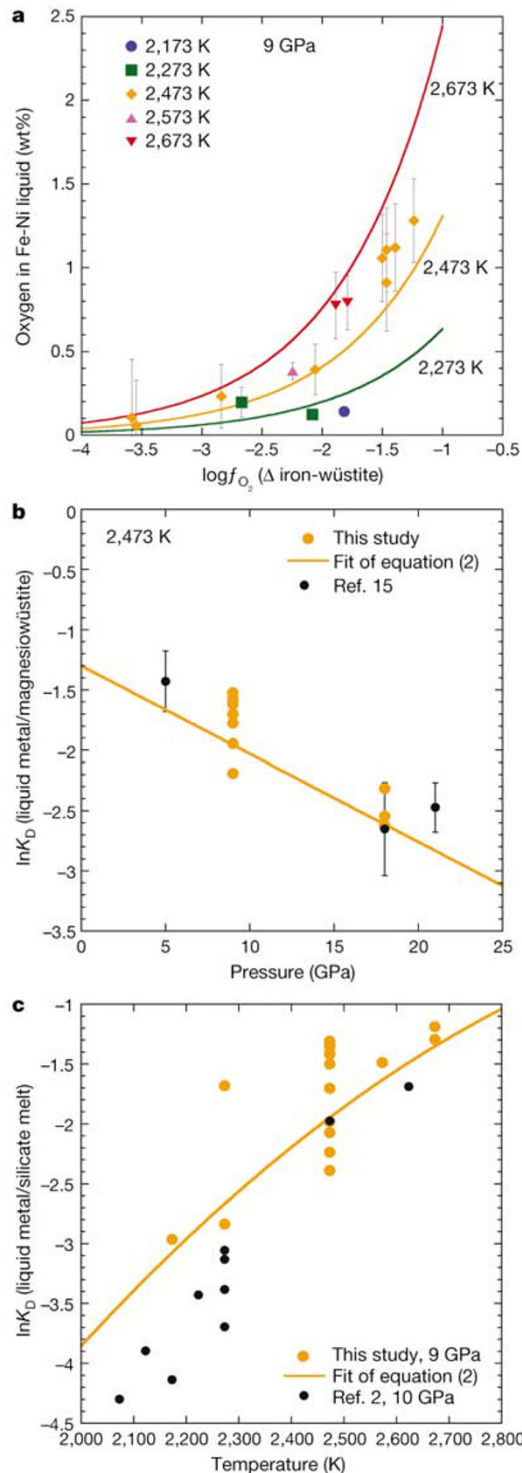


Figure 1 Oxygen partitioning results. **a**, **b**, Experimental results plotted as oxygen content in liquid Fe-alloy as a function of oxygen fugacity (f_{O_2}), relative to the iron-wüstite buffer, at 9 GPa and 2,173–2,673 K (**a**) and $\ln K_d$ as a function of pressure at 2,473 K; see equation (1) (**b**). At constant f_{O_2} , oxygen solubility increases with increasing temperature and decreases with increasing pressure. **c**, $\ln K_d$ for the partitioning of oxygen between liquid metal alloy and silicate melt calculated from liquid metal alloy/magnesiowüstite partitioning data, using equation (3). Values of $\ln K_d$ for oxygen partitioning between liquid Fe-Ni-S alloy and silicate melt, calculated from the results of ref. 2, are shown for comparison. The line is the fit of equation (2) to our data, modified using equation (3).

temperatures, using equation (2), are shown in Fig. 2. At moderate pressures (5–10 GPa) oxygen solubility is predicted to reach 5–15 wt% at 3,500–4,000 K. However, as pressure exceeds ~40 GPa, oxygen solubility becomes low, even at very high temperatures.

We examine the consequences of the partitioning of oxygen between liquid Fe-Ni metal and silicate liquid in a magma ocean. To determine the partitioning of oxygen between liquid metal and peridotite liquid in a magma ocean, we use the empirical relationship:

$$X_{\text{FeO}}^{\text{mw}} = 1.148X_{\text{FeO}}^{\text{sil}} + 1.319(X_{\text{FeO}}^{\text{sil}})^2 \quad (3)$$

where $X_{\text{FeO}}^{\text{sil}}$ is the mole fraction of FeO in silicate liquid. Equation (3) has been derived using the results of melting studies of peridotite and Fe-rich peridotite bulk compositions^{17,18}. Figure 1c shows the distribution coefficient K_d for the partitioning of oxygen between liquid Fe-alloy and silicate melt calculated from our liquid Fe-alloy/magnesiowüstite partitioning data using equation (3). These data and the recalculated fit of equation (2) are compared with previously published results on the partitioning of oxygen between liquid Fe-Ni-S alloy and silicate melt^{2,19}. The similar temperature dependence and absolute values of K_d observed indicates that the addition of up to 30 wt% sulphur has a very minor effect on oxygen solubility (Fig. 1c).

For a given bulk composition, the equilibrium compositions and mass fractions of coexisting metal and silicate are calculated by mass balance using equations (2) and (3). We consider the geochemical consequences of metal-silicate separation using a polybaric metal-silicate fractionation model²⁰. We assume that the temperature at the base of the magma ocean is defined by the peridotite liquidus^{21,22} and determine temperature as a function of depth by calculating magma ocean adiabats²⁰.

We assume that the material that accreted to form Earth and Mars contained the major elements Si, Al, Mg, Fe and Ca in chondritic (C1) proportions before metal-silicate separation²³. The oxygen contents of chondritic meteorites are variable and poorly defined²⁴ but, with the exception of enstatite chondrites, the silicate component is generally considerably more oxidized than the Earth's mantle. We therefore vary the amount of oxygen in the starting composition such that between 50 and 80 wt% of the total Fe is present initially as reduced metal, with the remainder being present as an FeO component. These two limits correspond to the non-metallic (silicate) fraction of the chondritic material containing 22 and 10 wt% FeO respectively (for comparison, the Earth's mantle contains ~8 wt% FeO).

The FeO content of the residual silicate mantle, after the metal has segregated, is calculated as a function of magma ocean depth for

Table 1 Experimental conditions and results

Run number	Temperature (K)	Pressure (GPa)	$\log(f_{O_2})$	Oxygen (wt%)	$\ln(K_d)$
1,549	2,173	9	-1.82 (0.01)	0.14 (0.01)	-3.218
1,558	2,273	9	-2.08 (0.01)	0.12 (0.01)	-3.077
1,358	2,273	9	-2.67 (0.09)	0.20 (0.09)	-1.903
1,203	2,473	9	-3.59 (0.35)	0.11 (0.35)	-1.525
1,186	2,473	9	-2.06 (0.15)	0.39 (0.15)	-1.945
1,316	2,473	9	-3.55 (0.27)	0.06 (0.27)	-2.189
1,210	2,473	9	-1.47 (0.29)	0.91 (0.29)	-1.776
1,218	2,473	9	-2.84 (0.19)	0.23 (0.19)	-1.574
1,383	2,473	9	-1.47 (0.25)	1.11 (0.25)	-1.585
1,194	2,473	9	-1.24 (0.25)	1.28 (0.25)	-1.704
1,371	2,473	9	-1.39 (0.26)	1.12 (0.26)	-1.7
1,352	2,473	9	-1.50 (0.26)	1.06 (0.26)	-1.62
1,547	2,573	9	-2.25 (0.05)	0.38 (0.05)	-1.721
1,312	2,673	9	-1.89 (0.20)	0.78 (0.20)	-1.435
1,551	2,673	9	-1.79 (0.16)	0.79 (0.16)	-1.549
1,273	2,473	18	-2.32 (0.20)	0.14 (0.20)	-2.621
1,275	2,473	18	-2.08 (0.18)	0.27 (0.18)	-2.314
1,285	2,473	18	-1.12 (0.25)	0.62 (0.25)	-2.545

A full list of sample analyses is provided in the Supplementary Information. $\log(f_{O_2})$ is calculated relative to the iron-wüstite buffer; K_d is defined in equation (1).

Earth and Mars (Fig. 3). In the case of Earth, the temperatures of magma oceans with depths <800 km are too low for significant amounts of oxygen to dissolve in the metal phase. Consequently, the initial FeO content of the silicate fraction of the chondritic material is changed only slightly by metal segregation (Fig. 3a). As the magma ocean depth is increased beyond 800 km, however, the amount of oxygen that partitions into the metal increases (owing to higher temperatures) with the consequence that the FeO content of the residual silicate mantle becomes increasingly reduced. Using the simple extrapolation model of equation (2), a magma ocean with a depth of ~1,800 km gives the current FeO content (8 wt%) and Fe/(Fe + Mg) ratio (0.1) of the Earth's mantle, assuming that the starting chondritic material was relatively oxidized with an initial FeO content of the order of 18–22 wt% (Fig. 3a). Considering the model uncertainties, the magma ocean depth could be as low as 1,200 km. A magma ocean with a depth of $\geq 1,200$ km has also been proposed on the basis of siderophile element partitioning studies^{2,25–27}.

In the case of Mars, the model predicts little change in the FeO content of the residual mantle as a result of core formation, irrespective of magma ocean depth (Fig. 3b). This is because the pressures and temperatures of a martian magma ocean are much lower than on Earth⁴; consequently, little or no oxygen dissolves in the liquid metal because of the low temperatures. Our results therefore show that Earth and Mars could have accreted from material with the same initial FeO content (~18 wt%) and that

the different FeO contents of their mantles are a consequence of the partitioning behaviour of oxygen during core formation. The model also explains the different mass fractions of the cores of Earth and Mars because more metal segregates from the magma ocean on Earth (~28 wt%) than on Mars (~18 wt%).

Although the model requires large extrapolations of the experimental data, the associated uncertainties do not affect our conclusions for Earth and Mars (see Supplementary Information). Using existing thermodynamic data^{28,29} on pure Fe liquid, oxygen and iron oxide we can calculate a value for ΔS that is in good agreement with our determined value. This calculation indicates that the inclusion of heat capacity functions to determine the temperature dependence of ΔS even up to 6,000 K has a very minor effect on the results of the model. The extrapolation assumes a constant positive value for ΔV of the metal–oxide equilibrium, which means that oxygen solubility decreases with pressure. This trend could reverse, however, at pressures higher than those of our experiments if the value for ΔV becomes negative as a result of compressibility or phase transformation³⁰. The martian mantle, however, extends only to pressures of 25 GPa and is therefore within

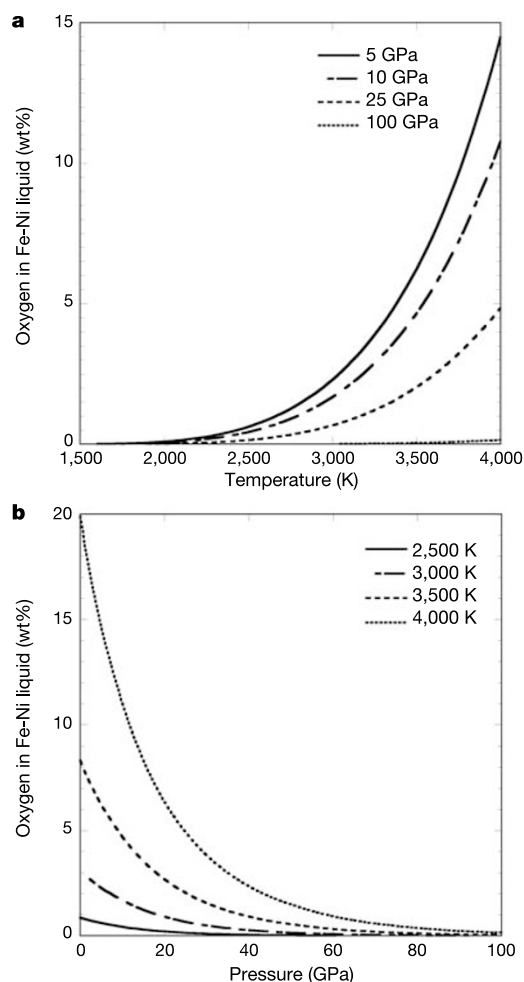


Figure 2 Extrapolations of the oxygen solubility data to higher temperatures and pressures (**a** and **b**) using equation (2). The oxygen fugacity is fixed at two log units below the iron–wüstite buffer.

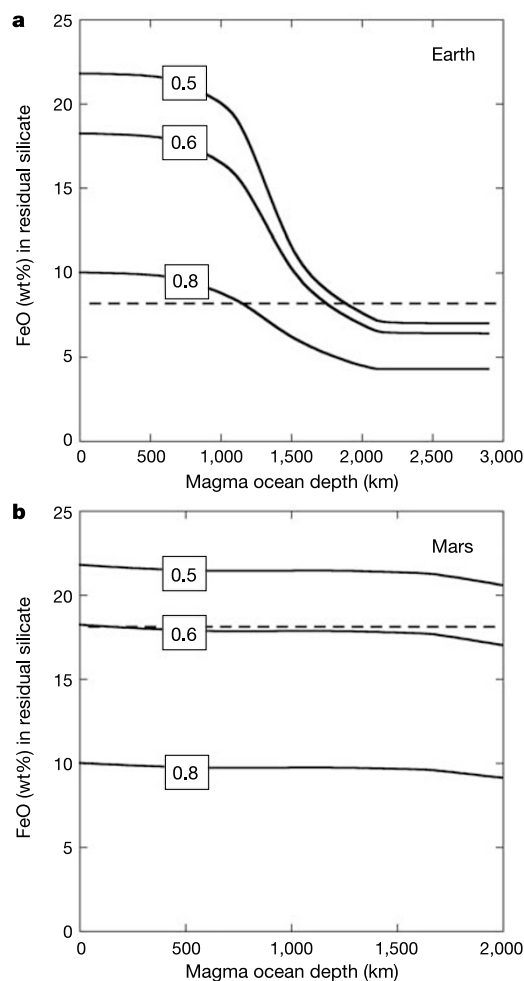


Figure 3 Results of the metal–silicate separation model for Earth (**a**) and Mars (**b**). The FeO contents of the residual silicate mantles (after metal segregation) are shown as a function of magma ocean depth. In each case, the three curves correspond to different initial oxygen contents and are labelled with the weight fraction of Fe that is initially present as metal; the three values correspond to 22, 18 and 10 wt% FeO in the silicate fraction, respectively. The results show that metal–silicate separation in a deep magma ocean would strongly reduce the FeO content of the silicate mantle on Earth, whereas on Mars metal–silicate separation has little effect. The horizontal broken lines indicate the FeO contents of the mantles of Earth and Mars.

the experimental pressure range. If oxygen solubility does increase at pressures higher than 25 GPa, this would reduce the magma ocean depth required on the Earth to satisfy the FeO content of the present-day mantle and make it more likely that oxygen remained dissolved in the metal as it formed the core.

An important consequence for the Earth is that the metal that segregated at the base of the magma ocean would have contained a significant amount of oxygen (for example, 7–8 wt%). As a result of increasing pressure, the solubility of oxygen in Fe-alloy is predicted to decrease with depth below the magma ocean (Fig. 2). There are three possibilities for the fate of the dissolved oxygen, all of which could be important. (1) FeO precipitated from liquid metal between the base of the magma ocean and the proto-core, thus creating chemical heterogeneities that may still exist in the lowermost mantle⁸. (2) Oxygen was transported by liquid metal to the core, where FeO precipitated to form the D'' layer at the core–mantle boundary⁹. (3) At least part of the oxygen may still be dissolved in the core (up to 8 wt% according to our model), thus contributing significantly to its light-element budget^{10,11}. If oxygen does become more soluble in liquid metal at pressures higher than our experimental conditions, then that would tend to favour this final possibility. In contrast, the martian core is unlikely to contain any dissolved oxygen, which is consistent with its probable high sulphur content⁷. □

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Convergent evolution in mechanical design of lamnid sharks and tunas

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The evolution of 'thunniform' body shapes in several different groups of vertebrates, including whales, ichthyosaurs¹ and several species of large pelagic fishes² supports the view that physical and hydromechanical demands provided important selection pressures to optimize body design for locomotion during vertebrate evolution. Recognition of morphological similarities between lamnid sharks (the most well known being the great white and the mako) and tunas has led to a general expectation that they also have converged in their functional design; however, no quantitative data exist on the mechanical performance of the locomotor system in lamnid sharks. Here we examine the swimming kinematics, *in vivo* muscle dynamics and functional morphology of the force-transmission system in a lamnid shark, and show that the evolutionary convergence in body shape and mechanical design between the distantly related lamnids and tunas is much more than skin deep; it extends to the depths of the myotendinous architecture and the mechanical basis for propulsive movements. We demonstrate that not only have lamnids and tunas converged to a much greater extent than previously known, but they have also developed morphological and functional adaptations in their locomotor systems that are unlike virtually all other fishes.

During their 400 million years of independent evolution, sharks and bony fishes have diverged in many fundamental aspects of their anatomy and physiology. However, two groups of dominant open-ocean predators, the lamnid sharks and the tunas, even when looked at superficially, display remarkably similar morphological specializations related to locomotion^{3–12} (Fig. 1a). The shared characteristics in these distantly related groups that distinguish them from virtually all other fish have arisen independently, probably as the result of selection for fast and continuous locomotion. Moreover, in both lamnids and tunas, the aerobic (red) musculature that powers cruise swimming is concentrated in a more medial (closer to the